

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
21 August 2003 (21.08.2003)

PCT

(10) International Publication Number  
**WO 03/069677 A1**

(51) International Patent Classification<sup>7</sup>: **H01L 29/12**,  
21/203, B82B 1/00, 3/00

DEN, Tohru [JP/JP]; c/o CANON KABUSHIKI  
KAISHA, 3-30-2, Shimomaruko, Ohta-ku, Tokyo  
146-8501 (JP).

(21) International Application Number: PCT/JP03/01368

(22) International Filing Date: 10 February 2003 (10.02.2003)

(74) Agents: OKABE, Masao et al.; No. 602, Fuji Bldg., 2-3,  
Marunouchi 3-chome, Chiyoda-ku, Tokyo 100-0005 (JP).

(25) Filing Language: English

(81) Designated States (*national*): AU, BR, CN, IN, KR, MX,  
SG, US.

(26) Publication Language: English

(30) Priority Data:  
2002-034006 12 February 2002 (12.02.2002) JP  
2002-340944 25 November 2002 (25.11.2002) JP

(84) Designated States (*regional*): European patent (AT, BE,  
BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU,  
IE, IT, LU, MC, NL, PT, SE, SI, SK, TR).

**Published:**

- with international search report
- before the expiration of the time limit for amending the  
claims and to be republished in the event of receipt of  
amendments

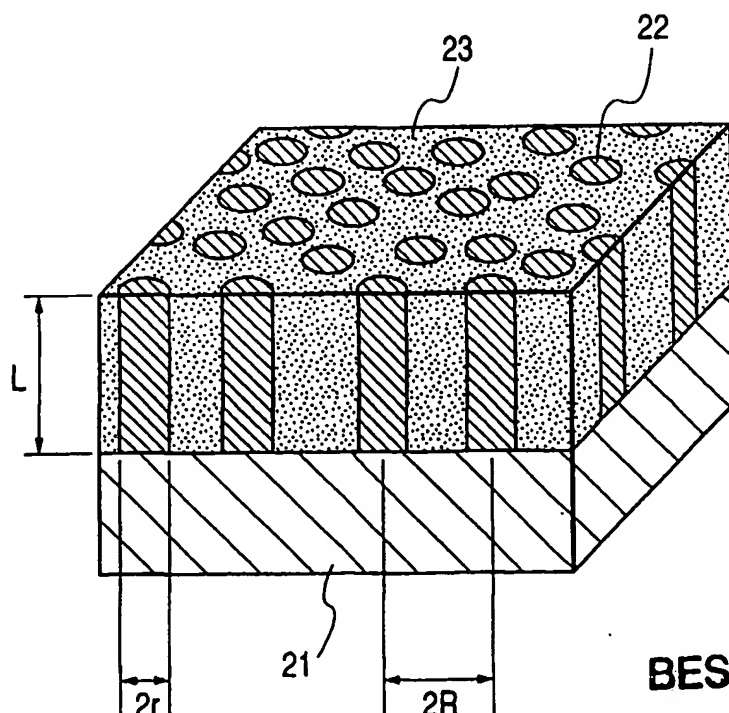
(71) Applicant (*for all designated States except US*): CANON  
KABUSHIKI KAISHA [JP/JP]; 3-30-2, Shimomaruko,  
Ohta-ku, Tokyo 146-8501 (JP).

(72) Inventors; and

(75) Inventors/Applicants (*for US only*): FUKUTANI,  
Kazuhiko [JP/JP]; c/o CANON KABUSHIKI KAISHA,  
3-30-2, Shimomaruko, Ohta-ku, Tokyo 146-8501 (JP).

*For two-letter codes and other abbreviations, refer to the "Guid-  
ance Notes on Codes and Abbreviations" appearing at the begin-  
ning of each regular issue of the PCT Gazette.*

(54) Title: STRUCTURE, METHOD OF MANUFACTURING THE SAME, AND DEVICE USING THE SAME



(57) Abstract: A composite structure is formed so as to contain aluminum and silicon or silicon / germanium. The composite structure comprises pillar-shaped members containing aluminum and a region containing silicon or silicon / germanium and surrounding the pillar-shaped members. The structure contains silicon or silicon / germanium at a content not less than 20 atomic% and not more than 70 atomic% relative to the total amount aluminum and silicon or silicon / germanium.

**BEST AVAILABLE COPY**

WO 03/069677 A1

## DESCRIPTION

STRUCTURE, METHOD OF MANUFACTURING THE SAME, AND  
DEVICE USING THE SAME

5

## TECHNICAL FIELD

This invention relates to a structure comprising pillar-shaped members, a method of manufacturing such a structure and a device using such a structure.

10

## BACKGROUND ART

Interests in microstructures have been growing in recent years from the viewpoint of utilizing them as functional materials.

15

Techniques that can be used for preparing such microstructures include those of directly preparing microstructures by utilizing semiconductor processing technologies, of which micro-pattern forming technologies such as photolithography are well known (see, inter alia, Japanese Patent Application Laid-Open No. 5-55545 (p. 3, FIG. 1)).

20

Apart from semiconductor processing technologies, techniques that utilize self-organization phenomena and self-formation phenomena of materials are also known. Such techniques are

25

devised to realize novel microstructures on the basis of regular structures that are formed in nature.

While a number of studies have been made on self-organization phenomena and self-formation phenomena because techniques that utilize such phenomena may feasibly be used to realize structures of dimensions of microns or even nanometers, there is still a strong demand for novel microstructures and methods of reliably manufacturing them.

#### DISCLOSURE OF THE INVENTION

In view of the above circumstances, it is therefore an object of the present invention to provide a novel structure, a method of manufacturing such a novel structure and a device that can be realized by utilizing such a novel structure.

In an aspect of the present invention, there is provided a structure containing a first material and a second material, the structure comprising pillar-shaped members containing the first material and a region containing the second material and surrounding the pillar-shaped members provided that the second material is not composed of only germanium, the structure containing the second

material at a content not less than 20 atomic% and not more than 70 atomic% relative to the total amount of the first and second materials.

5 The first material may typically be aluminum and the second material may typically be silicon or a mixture of silicon and germanium. The average diameter (or major axis if the pillar-shaped members show an elliptic plan view) of the pillar-shaped members may typically be not more than 50 nm  
10 and the gaps separating the centers of any adjacent ones of the pillar-shaped members contained in the structure is typically not more than 30 nm.

In another aspect of the present invention, there is provided a method of manufacturing a  
15 structure, comprising a step of preparing a substrate and a step of forming a film on the substrate in a non-equilibrium state, the film having a structure comprising pillar-shaped members containing a first material and a region containing  
20 a second material and surrounding the pillar-shaped members provided that the second material is not composed of only germanium, the structure containing the second material at a content not less than 20 atomic% and not more than 70 atomic%  
25 relative to the total amount of the first and second materials.

The present invention also provides a device

realized by comprising a structure according to the invention. For example, an electronic device according to the invention is realized by forming an insulated region on a structure according to the invention. An electronic device according to the invention may include single-electron transistors or single-electron memories or may include an information processing apparatus comprising such devices.

10           The present invention provides a film-shaped aluminum silicon composite structure, comprising pillar-shaped structures containing aluminum and a silicon region surrounding the pillar-shaped structures, the composite structure containing  
15           silicon at a content not less than 20 atomic% and not more than 70 atomic%.

          The present invention also provides a film-shaped aluminum silicon germanium composite structure, comprising pillar-shaped structures  
20           containing aluminum and a silicon germanium region surrounding the pillar-shaped structures, the composite structure containing silicon and germanium at a combined content not less than 20 atomic% and not more than 70 atomic%.

25           The present invention also provides a structure comprising a plurality of pillar-shaped members and a region surrounding the lateral

surfaces of the plurality of pillar-shaped members,  
the pillar-shaped members having a diameter not  
more than 20 nm, the distance between the centers  
of any two adjacently located ones of the pillar-  
5 shaped members being not more than 30 nm.

The present invention further provides a  
structure comprising pillar-shaped members  
containing aluminum and a region surrounding the  
pillar-shaped members, the pillar-shaped members  
10 and the region being formed simultaneously, the  
pillar-shaped members having a diameter not more  
than 20 nm.

A structure (e.g., aluminum silicon composite  
structure) comprising pillar-shaped members  
15 according to the invention may find applications as  
matrix that can be used for various devices. For  
example, it may find applications in various  
quantum devices including single-electron  
transistors and single-electron memories.

20

#### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1A is a schematic plan view of an  
embodiment of structure according to the invention;

FIG. 1B is a schematic cross sectional  
25 lateral view of the embodiment of FIG. 1A taken  
along line 1B-1B;

FIG. 2 is a schematic perspective view of a

structure according to the invention, illustrating a method of preparing it;

FIG. 3 is a schematic perspective view of another embodiment of structure according to the invention;

FIG. 4 is a schematic perspective view of still another embodiment of structure according to the invention;

FIG. 5 is a schematic cross sectional view of a single-electron transistor realized by using a structure according to the invention;

FIG. 6 is an FE-SEM image of an embodiment of aluminum silicon composite structure according to the invention;

FIG. 7 is an FE-SEM image of another embodiment of aluminum silicon composite structure according to the invention;

FIG. 8 is an FE-SEM image of still another embodiment of aluminum silicon composite structure according to the invention;

FIG. 9 is a schematic cross sectional view of a single-electron memory realized by using a structure according to the invention; and

FIG. 10 is an FE-SEM image of a composite structure according to the invention.

BEST MODE FOR CARRYING OUT THE INVENTION

Now, the present invention will be described in detail.

(Embodiment 1)

An embodiment of structure according to the invention will be described by referring to FIGS. 1A and 1B.

FIGS. 1A and 1B illustrate that pillar-shaped members 1 are distributed in a region 2 surrounding them. FIG. 1A is a schematic plan view of the embodiment and FIG. 1B is a schematic cross sectional view of the embodiment taken along line 1B-1B. In FIG. 1B, reference symbol 3 denotes a substrate.

The structure 100 illustrated in FIGS. 1A and 1B comprises pillar-shaped members 1 composed so as to contain a first material and surrounded by a region 2 composed so as to contain a second material. The structure 100 contains the second material at a content of not less than 20 atomic% and not more than 70 atomic% relative to the total amount of the first and second materials. As will be demonstrated hereinafter by referring to the succeeding embodiments, the inventors of the present invention found that it is possible to provide a structure comprising substantially pillar-shaped members distributed in a matrix region that surrounds them when the ratio of the



second material is within the above defined range relative to the total amount of the first material and the second material. If the substrate on which the structure is formed has a flat profile and is  
5 disposed horizontally, the pillar-shaped members are arranged generally vertically so as to stand upright, although the mode of arrangement of pillar-shaped members depends on the profile of the underlying substrate on which the structure is  
10 formed.

The above defined content refers to the ratio of the second material relative to the total amount of the first material and the second material of the structure. The content is preferably not less  
15 than 25 atomic% and not more than 65 atomic%, more preferably not less than 30 atomic% and not more than 60 atomic%. The content can be determined by quantitatively analyzing the structure, for example, by means of inductively coupled plasma atomic  
20 emission spectrometry (ICP-AES).

For the purpose of the present invention, it is necessary and sufficient that pillar-shaped members are substantially realized. The pillar-shaped members may contain the second material  
25 and/or the region may contain the first material. The pillar-shaped members and/or the surrounding region may contain impurities such as oxygen, argon,

etc.

Substances that can be used for the first material include Al (aluminum) and Au (gold).

Substances that can be used for the second material include Si (silicon), Ge (germinium), a mixture of Si and Ge (which may be expressed  $\text{Si}_x\text{Ge}_{1-x}$  ( $0 < x < 1$ ) hereinafter) and C. Particularly, it is preferable that the second material is selected from materials that can become amorphous.

10 The first and second materials preferably have an eutectic point in the phase equilibrium diagram of the binary system. Particularly, the eutectic temperature preferably is not lower than  $300^\circ\text{C}$ , more preferably not lower than  $400^\circ\text{C}$ .

15 Eutectoid materials may also be used as the first and second materials.

Preferable combinations of the first material and the second material include one using Al and Si respectively for the first and second materials and one using Al and  $\text{Si}_x\text{Ge}_{1-x}$  ( $0 < x < 1$ ) respectively for the first and second materials.

20 Preferably, the pillar-shaped members are at least partly polycrystalline and the region surrounding the pillar-shaped members is amorphous.

25 The pillar-shaped members show a circular or elliptic plan view.

Thus, in the structure, the pillar-shaped

members are distributed in the matrix composed so as to contain the second material. While the diameters (or major axes in the case where the pillar-shaped members do not show a circular plan view) of the pillar-shaped members can be controlled mainly by e.g. considering the composition of the structure (or the ratio of the second material), the average diameter is not less than 0.5 nm and not more than 50 nm, preferably not less than 1 nm and not more than 20 nm, more preferably not less than 2 nm and not more than 10 nm. The average diameter is most preferably less than 20 nm. The diameter as used herein refers to  $2r$  in FIG. 1B. In the case where the pillar-shaped members show an elliptic or other plan view, it is necessary and sufficient that the greatest axes corresponding to the diameters in the case of circular plan view are found within the above defined range. The average diameter refers to the value that is obtained from the images of the pillar-shaped members that can be observed in an SEM photograph (of an area of about  $100\text{ nm} \times 100\text{ nm}$ ) directly or by processing the photograph by means of a computer. It should be noted here that the optimum values of the diameter and the gap as defined below can vary depending on the materials and the composition involved and the application of

the structure.

The distance  $2R$  between the centers of any two adjacently located pillar-shaped members (FIG. 1B) is not less than 2 nm and not more than 30 nm, preferably not less than 5 nm and not more than 20 nm, more preferably not less than 5 nm and not more than 15 nm. Of course, the smallest distance  $2R$  between the centers of any two adjacently located pillar-shaped members needs to be such that the two pillar-shaped structures do not contact each other. If the second material (the material contained in the region surrounding the pillar-shaped members) is a mixture of a plurality of elements (e.g.,  $\text{Si}_x\text{Ge}_{1-x}$ ), the distance between the centers of any two adjacently located pillar-shaped members can be controlled by way of the mixing ratio of elements.

Preferably, the structure is a film-shaped structure. If such is the case, the pillar-shaped members are distributed in the matrix composed so as to contain the second material in such a way that they are substantially perpendicular relative to the intra-surface direction. While the film thickness of a film-shaped structure is not particularly limited, it may normally be between 1 nm and 100  $\mu\text{m}$ . More preferably, it may be between 1 nm and 1  $\mu\text{m}$  or between 1 nm and 3  $\mu\text{m}$  from a realistic viewpoint of considering the process time.

Particularly, it is preferable that the pillar-shaped structures are maintained even when the film thickness is not less than 300 nm. The pillar-shaped members preferably do not substantially have  
5 any branches extending to upwardly or downwardly (in the longitudinal direction of the pillar-shaped members).

As pointed out above, the structure is preferably a film-shaped structure and may be  
10 arranged on a substrate. When a substrate is used, it is not subjected to any particular limitations. Substrates that can suitably be used for the purpose of the invention include insulator substrates such as quartz glass substrates,  
15 reinforced glass substrates, crystallized glass substrates or other glass substrates, substrates having an insulating layer on the surface, semiconductor substrates including silicon substrates (including p-type, n-type, high  
20 resistance type, low resistance type, etc.), gallium substrates and indium phosphide substrates, metal substrates including aluminum substrates and flexible substrates (e.g., polyimide resin substrates) provided that the substrate operates as  
25 support member and a structure according to the invention can be formed on it.

It is possible to provide an electronic

device by forming an insulating film on a structure according to the invention. The expression of an electronic device as used herein refers to a single-electron transistor, a single-electron memory or the like. For the purpose of the invention, an electronic device may be an information processing apparatus comprising such devices.

A structure according to the invention (which may also be referred to as composite structure hereinafter) may find applications as matrix that can be used for various devices. For example, it may find applications in various quantum devices including single-electron transistors and single-electron memories and also in micro-electrodes, etc.

A structure according to the invention (e.g., an aluminum silicon composite structure) can be used as dry or wet etching mask for the purpose of processing some other substrate or a film formed on some other substrate.

According to the invention, pillar-shaped structures such as quantum dots or quantum wires can be utilized in various different ways to remarkably broaden the scope of utilization of such structures. A structure according to the invention can be used as self-functioning material by itself.

(Manufacturing Method)

A structure according to the invention can be prepared by utilizing a method of forming a film in a non-equilibrium state. While a sputtering method is preferably used for forming a film for the purpose of the invention, some other film forming method adapted to form a film in an appropriately selected non-equilibrium state such as a resistance heating evaporation method, an electron beam evaporation (EB evaporation) method or an ion plating method may alternatively be used. If a sputtering method is used, it may be a magnetron sputtering method, an RF sputtering method, an ECR sputtering method or a DC sputtering method. When a film forming process is conducted by using a sputtering method, it is conducted in a reaction furnace containing an argon atmosphere whose internal pressure is held to a level between 0.2 and 1 Pa or between 0.1 and 1 Pa. While the first material and the second material may be used separately as target materials for the sputtering operation (e.g., as separate aluminum and silicon target materials), a target material obtained by sintering the first material and the second material that are mixed to a desired ratio may alternatively be used. For this embodiment of structure, pillar-shaped members and a region surrounding the lateral surfaces of the pillar-

shaped members are formed simultaneously.  
Preferably, the sputtering operation is conducted without bringing plasma into contact with the substrate on which a film is grown.

5           A structure according to the invention can maintain a state where the members having a pillar-shaped structure are distributed in a region surrounding the lateral surfaces of the members even if the film is formed to a thickness greater  
10           than a predetermined value. In other words, even if the film thickness is unexpectedly large, the diameter of the pillar-shaped structures would not vary significantly. The predetermined film  
            thickness is not less than 110 nm, preferably not  
15           less than 300 nm.

            The structure is formed on a substrate whose temperature is not higher than 300°C, preferably not lower than 20°C and not higher than 200°C, more preferably not lower than 100°C and not higher than  
20           150°C.

(Embodiment 2: aluminum silicon composite structure)

            A structure containing aluminum and silicon respectively as the first and second materials will  
25           be described also by referring to FIGS. 1A and 1B.

            FIG. 1A is schematic plan view of an aluminum silicon composite structure according to the



invention. FIG. 1B is a schematic cross sectional view of the specimen taken along line 1B-1B in FIG. 1A. In FIGS. 1A and 1B, reference symbols 1 and 2 respectively denote pillar-shaped structures including aluminum and a region surrounding the pillar-shaped structures. In FIG. 1B, reference symbol 3 denotes a substrate.

The aluminum silicon composite structure 100 formed on the substrate 3 contains silicon at a content not less than 20 atomic% and not more than 70 atomic%, preferably not less than 25 atomic% and not more than 65 atomic%, more preferably not less than 30 atomic% and not more than 60 atomic%, relative to the total amount of aluminum and silicon. An aluminum silicon composite structure in which pillar-shaped structures 1 are distributed in a silicon region 2 is obtained when the ratio of silicon is found within the above defined range.

The content (atomic%) represents a value obtained by quantitatively analyzing the amount of silicon and that of aluminum in the film-shaped aluminum silicon composite structure by means of inductively coupled plasma atomic emission spectrometry.

While atomic% is used above as unit for expressing the content, wt% may alternatively be used as unit. If such is the case, the expression

of not less than 20 atomic% and not more than 70 atomic% is replaced by an expression of not less than 20.65 wt% and not more than 70.84 wt% (obtained as a result of reductions using 26.982 and 28.086 for the atomic amounts of Al and Si respectively).

An aluminum silicon composite structure 100 according to the invention comprises aluminum pillar-shaped structures containing aluminum as principal ingredient and a silicon region surrounding them and containing silicon as principal ingredient.

While the pillar-shaped structure section 1 contains aluminum as principal ingredient, they may additionally contain silicon, oxygen and/or argon as other ingredients so long as a microstructure having pillar-shaped structures is obtained. The expression of principal ingredient as used herein refers to the composition of the pillar-shaped structure section where the aluminum content is not less than 50 atomic%, preferably not less than 80 atomic%.

While the silicon region that surrounds the pillar-shaped structures contains silicon as principal ingredient, it may additionally contain aluminum, oxygen and/or argon as other ingredients so long as a microstructure having pillar-shaped

structures is obtained. The expression of principal ingredient as used herein refers to the composition of the silicon region where the silicon content is not less than 50 atomic%, preferably not less than 80 atomic%.

Desirably, the silicon region is amorphous. From the viewpoint of insulating effect, it is desirable that the silicon region is made of amorphous silicon because a higher electric insulating effect of the matrix material separating the pillar-shaped structures are obtained due to a greater band gap and a higher defect density when amorphous silicon is used than when crystalline silicon is used.

A composite structure according to the invention is in a state where aluminum is isolated in a silicon matrix.

(Structure)

The pillar-shaped structures 1 that contain aluminum usually show a circular or elliptic shape as viewed in plan. Of course, they may show any other plan view so long as they are appropriately distributed in the silicon region 2.

While there are no particular limitations to the diameter of the pillar-shaped structures of an aluminum silicon composite structure according to the invention, the average diameter is not less

than 0.5 nm and not more than 50 nm, preferably not less than 0.5 nm and not more than 20 nm, more preferably not less than 0.5 nm and not more than 10 nm. The diameter as used herein refers to  $2r$  in FIG. 1B. In the case where the pillar-shaped structures show an elliptic or other plan view, it is necessary and sufficient that the greatest axes corresponding to the diameters in the case of circular plan view are found within the above defined range. The average diameter refers to the value that is obtained from the images of the pillar-shaped structures of aluminum that can be observed in an SEM photograph (of an area of about  $100\text{ nm} \times 100\text{ nm}$ ) by processing the photograph by means of a computer.

Meanwhile, a nano-structure having a diameter of nanometers (in a range between about 0.1 nm and 100 nm) can show peculiar electric, optical and chemical properties when the dimensions are made smaller than a characteristic value because movements of electrons are confined in such a condition. Thus, a nano-structure that is used as functional material can be very useful from this viewpoint. An aluminum silicon composite structure according to the invention can be utilized in various different ways as nano-structure when the pillar-shaped structures it comprises show a

diameter not less than 0.5 nm and not more than 50 nm, particularly not less than 0.5 nm and not more than 10 nm.

The distance  $2R$  between the centers of any two adjacently located pillar-shaped structures (FIG. 1B) is not more than 30 nm, preferably not more than 15 nm. Of course, the distance  $2R$  between the centers of any two adjacently located pillar-shaped structures needs to be such that the two pillar-shaped structures do not contact each other. Particularly, it is desirable that both the average diameter  $2r$  of the pillar-shaped structures and the distance  $2R$  between the centers of any two adjacently located pillar-shaped structures are found within the above defined respective ranges.

A typical microstructure according to the invention comprises aluminum nano-structures having a diameter between 1 and 9 nm, the distance between the centers of any two adjacently located aluminum nano-structures being between 5 and 10 nm, the ratio of the height to the diameter of the aluminum nano-structures being between 0.1 and 100,000, the aluminum nano-structures being arranged perpendicularly relative to the substrate.

As illustrated in FIG. 1B, the pillar-shaped structures 1 may preferably show a rectangular cross sectional view taken through the substrate.

Alternatively, they may show a square or trapezoidal cross sectional view. The pillar-shaped structures may show a profile with an appropriate aspect ratio (length / diameter). The aspect ratio (length  $L$  / diameter  $2r$ ) may be  
5 between 0.1 and 100,000.

For example, the length  $L$  of the pillar-shaped structures may be between 1 nm and 100  $\mu\text{m}$ .

Particularly, assuming that the diameter  $2r$  of the pillar-shaped structures is between 1 and 10  
10 nm and the distance  $2R$  between the centers of any two adjacently located pillar-shaped structures is between 5 and 15 nm, the length  $L$  can be controlled so as to be between 1 nm and several  $\mu\text{m}$  in a manner as described below. The pillar-shaped structures 1  
15 operate as aluminum quantum dots when the length  $L$  is between several nm and tens of several nm (and hence the ratio of the length to the diameter is small), whereas they operate as aluminum quantum  
20 wires when the length  $L$  is greater.

As shown in FIG. 1B, the pillar-shaped structures 1 that contain aluminum are separated from each other by the silicon region that contains silicon as principal ingredient. In other words, a  
25 number of pillar-shaped structures are distributed in the silicon region.

Preferably, the pillar-shaped structures 1

that contain aluminum are arranged in a specific direction. Particularly, they are arranged in a direction perpendicular to the substrate as shown in FIG. 1B.

5           While the substrate 3 is not subjected to particular limitations, it may be selected from insulator substrates such as quartz glass substrates, semiconductor substrates including silicon substrates, gallium arsenide substrates and  
10   indium phosphide substrates, and flexible substrates (e. g., polyimide resin substrates) provided that the substrate operates as support member and an aluminum silicon composite structure according to the invention can be formed on it.  
15   Further, substrates on which one or more than one film layers are formed may also be used.  
(Embodiment 3: method of preparing an aluminum silicon composite structure)

          Now, a method of preparing an aluminum  
20   silicon composite structure according to the invention will be described by referring to FIG. 2. More specifically, a sputtering method is used for preparing an aluminum silicon composite structure as exemplary method for forming a film in a non-  
25   equilibrium state in the following description. In FIG. 2, reference symbols 11 and 12 respectively denote a substrate and a sputtering target. The

ratio of the aluminum content to the silicon content can be easily modified when a sputtering method is used.

As shown in FIG. 2, an aluminum silicon  
5 composite film is formed on a substrate by means of a magnetron sputtering method that is a method for forming a film in a non-equilibrium state.

Raw materials of silicon and aluminum are provided properly by arranging silicon chips on an  
10 aluminum target substrate as shown in FIG. 2. While a number of silicon chips are arranged on the aluminum target substrate in FIG. 2, the arrangement of silicon is not limited to it. Only a single silicon chip may alternatively be arranged  
15 on the aluminum target substrate so long as a desired film is formed. Note however, silicon chips are preferably arranged symmetrically on the substrate 11 if aluminum-containing pillar-shaped structures are to be uniformly distributed in a  
20 silicon region.

It is also possible to use a baked aluminum / silicon body prepared by baking a predetermined amount of powder of aluminum and silicon as target.

Similarly, it is also possible to perform a  
25 sputtering process simultaneously on an aluminum target and a silicon target that are prepared separately.



The silicon content of the film that is produced by sputtering is not less than 20 atomic% and not more than 70 atomic%, preferably not less than 25 atomic% and not more than 65 atomic%, more preferably not less than 30 atomic% and not more than 60 atomic% relative to the total amount of aluminum and silicon.

The substrate temperature is not higher than 300°C, preferably not lower than 20°C and not higher than 200°C, more preferably not lower than 100°C and not higher than 150°C. If the silicon content is found within the above defined range, an aluminum silicon composite structure comprising pillar-shaped structures distributed in a silicon region is obtained.

The temperature of the specimen of aluminum silicon composite film being formed is typically not higher than 300°C, preferably not higher than 200°C. When a composite film of aluminum and silicon is formed by means of a method of forming a film of substances in a non-equilibrium state at specimen temperature not higher than 300°C, the prepared aluminum silicon composite film shows an eutectic composition where aluminum and silicon are in a metastable state and aluminum forms pillar-shaped nano-structures of dimensions of several nm and becomes separated in a self-forming manner.

The silicon content of an aluminum silicon composite structure according to the invention can be controlled typically by arranging an appropriate amount of silicon chips on an aluminum target.

5           When a film is formed in a non-equilibrium state particularly by sputtering, the pressure in the reactor in which argon gas is made to flow is preferably between 0.2 and 1 Pa or between 0.1 and 1 Pa, although the pressure is by no means limited  
10       to the above range so long as argon plasma is produced on a stable basis under that pressure.

          The substrate 11 may be selected from insulator substrates such as quartz glass substrates, semiconductor substrates including  
15       silicon substrates and gallium arsenide substrates and substrates on which one or more than one film layers are formed. The material, the thickness and the mechanical strength of the substrate are not subjected to any particular limitations so long as  
20       it does not provide any problem for forming an aluminum silicon nano-structure. The profile of the substrate is not limited to that of a smooth and flat plate. In other words, the surface of the substrate may be curved or have undulations or  
25       steps to a certain extent. Thus, there are no particular limitations to the profile of the substrate so long as a desired aluminum silicon

nano-structure is formed properly.

The method of forming a film of substances in a non-equilibrium state to be used for this embodiment is preferably a sputtering method, although some other appropriate method of forming a film of substances in a non-equilibrium state may be used. More specifically, a resistance heating evaporation method, an electron beam evaporation (EB evaporation) method or an ion plating method may be used for this embodiment.

For forming a film, a simultaneous film forming process of forming a film of silicon and aluminum at the same time may be used. Alternatively, a multilayer film forming process of forming several atomic layers of silicon and those of aluminum successively may be used. (Embodiment 4: device comprising an aluminum silicon composite structure)

FIG. 5 is a schematic cross sectional view of a single-electron transistor realized by using an aluminum silicon composite structure according to the invention. As aluminum is used as quantum dots as shown in FIG. 5, the electric current flowing between the source and the drain can be made on and off by controlling the voltage applied to the gate electrode. Thus, the device of FIG. 5 operates as transistor. In FIG. 5, there are shown a substrate

51, an insulating body (e.g., silicon oxide body)  
52, an aluminum silicon composite structure 53, a  
drain electrode 54, a gate insulator (insulating  
film) 55, a gate electrode 56 and a source  
5 electrode 57. Of course, a composite structure  
made of materials other than aluminum and silicon  
may be used in place of the aluminum silicon  
composite structure 53 so long as such a structure  
comprises pillar-shaped structures. More  
10 specifically, silicon of the aluminum silicon  
composite structure 53 may be replaced by silicon  
and germanium so long as a structure comprising  
distributed pillar-shaped structures are realized.

A single-electron memory that utilizes the  
15 quantum effect, or a single-electron transistor,  
can be formed by using an aluminum silicon  
structure prepared in a manner as described above.

According to the invention, aluminum nano-  
structures such as quantum dots or quantum wires  
20 can be utilized in various different ways to  
remarkably broaden the scope of utilization of such  
structures. A structure according to the invention  
can be used as self-functioning material by itself.  
(Embodiment 5: the composition of an aluminum  
25 silicon germanium composite structure)

The use of aluminum and  $\text{Si}_x\text{Ge}_{1-x}$  ( $0 < x < 1$ )  
respectively as the first material for forming

pillar-shaped members and as the second material will be described by referring to FIGS. 1A and 1B again.

FIG. 1A is schematic plan view of an aluminum silicon germanium composite structure according to the invention. FIG. 1B is a schematic cross sectional view of the specimen taken along line 1B-1B in FIG. 1A. In FIGS. 1A and 1B, reference symbols 1 and 2 respectively denote aluminum-containing pillar-shaped structures and a silicon germanium region surrounding the pillar-shaped structures. In FIG. 1B, reference symbol 3 denotes a substrate.

An aluminum silicon germanium composite structure 100 formed on the substrate 3 contains silicon and germanium, the combined content of silicon and germanium being not less than 20 atomic% and not more than 70 atomic%, preferably not less than 25 atomic% and not more than 65 atomic%, more preferably not less than 30 atomic% and not more than 60 atomic%, relative to the total amount of aluminum, silicon and germanium. An aluminum silicon germanium composite structure in which pillar-shaped structures 1 are distributed in a silicon germanium region 2 is obtained when the ratio of silicon germanium is found within the above defined range.

The combined content (atomic%) represents a value obtained by quantitatively analyzing the amount of silicon and germanium and that of aluminum in the film-shaped aluminum silicon germanium composite structure by means of inductively coupled plasma atomic emission spectrometry.

The combined content of silicon and germanium relative to the total amount of aluminum, silicon and germanium is expressed by  $(\text{Si} + \text{Ge}) / (\text{Si} + \text{Ge} + \text{Al}) \times 100$ , where Si is the silicon content, Ge is the germanium content and Al is the aluminum content. In other words, it is the combined content of Si + Ge expressed in terms of atomic% when Si + Ge + Al is 100 atomic%.

The content ratio of silicon to germanium in the silicon germanium region of an aluminum silicon germanium nano-structure according to the invention is not particularly limited. In other words, it is necessary and sufficient that the silicon germanium region contains at least both silicon and germanium. If the content ratio of silicon (Si) to germanium (Ge) is expressed by  $\text{Si}_x\text{Ge}_{1-x}$ , it is necessary and sufficient that  $0 < x < 1$ . When the content ratio is found within the above range, it is possible to control the diameters of or gaps separating the aluminum pillar-shaped nano-structures of an

aluminum silicon germanium nano-structure according to the invention better than the diameters of or gaps separating the aluminum pillar-shaped nano-structures of an aluminum silicon nano-structure or an aluminum germanium nano-structure. From this point of view, the present invention includes a method of controlling the diameters of or gaps separating the pillar-shaped members of a structure according to the invention.

10           An aluminum silicon germanium composite structure according to the invention comprises aluminum pillar-shaped structures containing aluminum as principal ingredient and a silicon germanium region surrounding them and containing  
15           silicon and germanium as principal ingredient.

          While the pillar-shaped structure section 1 contains aluminum as principal ingredient, they may additionally contain silicon, germanium, oxygen and/or argon as other ingredients so long as a  
20           microstructure having pillar-shaped structures is obtained. The expression of principal ingredient as used herein refers to the composition of the pillar-shaped structure section where the aluminum content is not less than 50 atomic%, preferably not  
25           less than 80 atomic%.

          While the silicon germanium region that surrounds the pillar-shaped structures contains

silicon and germanium as principal ingredient, it may additionally contain aluminum, oxygen and/or argon as other ingredients so long as it surrounds pillar-shaped microstructures containing aluminum.

5 The expression of principal ingredient as used herein refers to the composition of the silicon germanium region where the combined content of silicon and germanium is not less than 50 atomic%, preferably not less than 80 atomic%.

10 Desirably, the silicon germanium region is amorphous. From the viewpoint of insulating effect, it is desirable that the silicon germanium region is made of amorphous silicon germanium because a higher electric insulating effect of the matrix  
15 material separating the pillar-shaped structures are obtained due to a greater band gap and a higher defect density when amorphous silicon germanium is used than when crystal silicon germanium is used.

A composite structure according to the  
20 invention is in a state where aluminum is isolated in a silicon germanium matrix.

(Structure)

The pillar-shaped structures 1 that contain aluminum show a circular or elliptic plan view. Of  
25 course, they may show any other plan view so long as they are appropriately distributed in the silicon germanium region 2.



While there are no particular limitations to the diameter of the pillar-shaped structures of an aluminum silicon germanium composite structure according to the invention, the average diameter is  
5 not less than 0.5 nm and not more than 30 nm, preferably not less than 0.5 nm and not more than 20 nm, more preferably not less than 0.5 nm and not more than 15 nm. The lower limit may be 1 nm or 2 nm. The diameter as used herein refers to  $2r$  in  
10 FIG. 1B. In the case where the pillar-shaped members show an elliptic or other plan view, it is necessary and sufficient that the greatest axes corresponding to the diameters in the case of circular plan view are found within the above  
15 defined range. The average diameter refers to the value that is obtained from the images of the pillar-shaped members of aluminum that can be observed in an SEM photograph (of an area of about  $100\text{ nm} \times 100\text{ nm}$ ) by processing the photograph by  
20 means of a computer. The aluminum part is assumed to show an elliptic plan view and the average value of major axes are calculated.

Meanwhile, a nano-structure having a diameter (major axis) of nanometers (in a range between  
25 about 0.1 nm and 100 nm) can show peculiar electric, optical and chemical properties when the dimensions are made smaller than a characteristic value (e.g.,

mean free path). Thus, a nano-structure that is used as functional material can be very useful from this viewpoint. An aluminum silicon germanium composite structure according to the invention can be utilized in various different ways as nano-structure when the pillar-shaped structures it comprises show a diameter not less than 0.5 nm and not more than 30 nm, particularly not less than 0.5 nm and not more than 15 nm.

10           The distance  $2R$  between the centers of any two adjacently located pillar-shaped structures 1 (Fig. 1B) is not more than 30 nm, preferably not more than 20 nm. Of course, the distance  $2R$  between the centers of any two adjacently located pillar-shaped structures needs to be such that the two pillar-shaped structures do not contact each other. Particularly, it is desirable that both the average diameter  $2r$  of the pillar-shaped structures and the distance  $2R$  between the centers of any two adjacently located pillar-shaped structures are found within the above defined respective ranges.

20           A typical microstructure according to the invention comprises pillar-shaped aluminum nano-structures having a diameter between 1 and 15 nm, the distance between the centers of any two adjacently located aluminum nano-structures being between 10 and 20 nm, the ratio of the height to

the diameter of the aluminum nano-structures being between 0.1 and 100,000, the aluminum nano-structures being arranged perpendicularly relative to the substrate.

5           As illustrated in FIG. 1B, the pillar-shaped structures 1 may show a rectangular cross sectional view taken through the substrate. Alternatively, they may show a square or trapezoidal cross sectional view. The pillar-shaped structures may  
10 show a profile with an appropriate aspect ratio (length / diameter). The aspect ratio (length  $L$  / diameter  $2r$ ) may be between 0.1 and 100,000.

For example, the length  $L$  of the pillar-shaped structures may be between 1 nm and 100  $\mu\text{m}$ .

15           Particularly, assuming that the diameter  $2r$  of the pillar-shaped structures is between 1 and 15 nm and the distance  $2R$  between the centers of any two adjacently located pillar-shaped structures is between 10 and 20 nm, the length  $L$  can be  
20 controlled so as to be between 1 nm and several  $\mu\text{m}$  in a manner as described below. The pillar-shaped structures 1 operate as aluminum quantum dots (zero-dimensional) when the length  $L$  is between several nm and tens of several nm (and hence the  
25 ratio of the length to the diameter is small), whereas they operate as aluminum quantum wires (one-dimensional) when the length  $L$  is greater.

As shown in FIG. 1B, the pillar-shaped structures 1 that contain aluminum are separated from each other by the silicon germanium region that contains silicon and germanium as principal ingredient. In other words, a number of pillar-shaped structures are distributed in the silicon germanium region.

Preferably, the pillar-shaped structures 1 that contain aluminum are arranged in a specific direction. Particularly, they are arranged in a direction perpendicular to the substrate as shown in FIG. 1B.

While the substrate 3 is not subjected to particular limitations, it may be selected from insulator substrates such as quartz glass substrates and plastic substrates, semiconductor substrates including silicon substrates, germanium substrates, gallium arsenide substrates and indium phosphide substrates and flexible substrates (e.g., polyimide resin substrates) provided that the substrate operates as support member and an aluminum silicon germanium structure according to the invention can be formed on it. Substrates on which one or more than one film layers formed may also be used.

(Embodiment 6: method of preparing an aluminum silicon germanium composite structure)

Now, a method of preparing an aluminum silicon germanium composite structure according to the invention will be described by also referring to FIG. 2. More specifically, a sputtering method is used for preparing an aluminum silicon germanium composite structure as exemplary method for forming a film in a non-equilibrium state in the following description. In FIG. 2, reference symbols 11 and 12 respectively denote a substrate and a sputtering target. The ratio of the contents of aluminum, silicon and germanium can be easily modified by regulating the target materials when a sputtering method is used. In this embodiment, reference symbol 13 denotes silicon chips or germanium chips.

As shown in FIG. 2, an aluminum silicon germanium composite film is formed on a substrate by means of a magnetron sputtering method that is a method for forming a film in a non-equilibrium state.

Raw materials of silicon, germanium and aluminum are provided properly by arranging silicon chips and germanium chips on an aluminum target substrate as shown in FIG. 2. While a number of silicon chips and germanium chips are arranged on the aluminum target substrate in FIG. 2, the arrangement of silicon or germanium chips is not limited to it. Only a single silicon chip or

germanium chip may alternatively be arranged on the aluminum target substrate so long as a desired film is formed. Note however, silicon chips and germanium chips are preferably arranged

5     symmetrically on the substrate 11 if aluminum-containing pillar-shaped structures are to be uniformly distributed in a silicon germanium region.

It is also possible to use aluminum silicon germanium sintered body prepared by baking a

10     predetermined amount of powder of aluminum, silicon and germanium as target for forming a film. A homogeneous film that is practically free from variances of film composition can be obtained by using such a target.

15     Similarly, it is also possible to perform a sputtering process simultaneously on an aluminum target, a silicon target and a germanium target that are prepared separately.

The combined content of silicon and germanium

20     of the film that is produced by sputtering is not less than 20 atomic% and not more than 70 atomic%, preferably not less than 25 atomic% and not more than 65 atomic%, more preferably not less than 30 atomic% and not more than 60 atomic% relative to

25     the total amount of aluminum, silicon and germanium.

The substrate temperature is not higher than 300°C, preferably not higher than 200°C, more

preferably not lower than 100°C and not higher than 150°C. The lower limit is 0°C or room temperature. If the content of silicon and germanium is found within the above defined range, an aluminum silicon  
5 germanium composite structure comprising pillar-shaped structures distributed in a silicon germanium region is obtained at substrate temperature within the above range.

The temperature of the specimen of aluminum  
10 silicon germanium composite film being formed is typically not higher than 300°C, preferably not higher than 200°C. When a composite film of aluminum, silicon and germanium is formed by means of a method of forming a film of substances in a  
15 non-equilibrium state at specimen temperature not higher than 300°C though depending on film forming conditions, the prepared aluminum silicon germanium composite film shows an eutectic composition where aluminum, silicon and germanium are in a metastable  
20 state and aluminum forms pillar-shaped nano-structures of dimensions of several nm and becomes separated from the silicon germanium region in a self-forming manner.

The combined content of silicon and germanium  
25 in an aluminum silicon germanium composite structure according to the invention can be controlled typically by arranging an appropriate

amount of silicon chips or germanium chips on an aluminum target or by using a target prepared by using an appropriate mixing ratio of aluminum, silicon and germanium in a powdery state.

5           When a film is formed in a non-equilibrium state particularly by sputtering, the pressure in the reactor in which argon gas is made to flow is preferably between 0.2 and 1 Pa or between 0.1 and 1 Pa, although the pressure is by no means limited  
10 to the above range so long as argon plasma is produced on a stable basis under that pressure.

          The substrate 11 may be selected from insulator substrates such as quartz glass  
substrates, semiconductor substrates including  
15 silicon substrates and gallium arsenide substrates and substrates on which one or more than one film layers are formed. The material, the thickness and the mechanical strength of the substrate are not  
subjected to any particular limitations so long as  
20 it does not provide any problem for forming an aluminum pillar-shaped nano-structure. The profile of the substrate is not limited to that of a smooth and flat plate. In other words, the surface of the substrate may be curved or have undulations or  
25 steps to a certain extent. Thus, there are no particular limitations to the profile of the substrate so long as a desired aluminum pillar-



shaped nano-structure is formed properly.

The method of forming a film of substances in a non-equilibrium state to be used for this embodiment is preferably a sputtering method, although some other appropriate method of forming a film of substances in a non-equilibrium state may be used. More specifically, an evaporation method (resistance heating evaporation, electron beam evaporation, etc.), an ion plating method or some other method that can form a film of substances in a non-equilibrium state may be used for this embodiment.

For forming a film, a simultaneous film forming process of forming a film of silicon, germanium and aluminum at the same time may be used. Alternatively, a multilayer film forming process of successively forming several atomic layers of silicon, those of germanium and those of aluminum may be used.

(Embodiment 7: device comprising an aluminum silicon germanium composite structure)

FIG. 9 is a schematic cross sectional view of a single-electron memory realized by using an aluminum silicon germanium composite structure according to the invention.

In FIG. 9, there are shown a substrate 91, an insulating body (e.g., silicon oxide body) 92, a

structure 93 in which pillar-shaped members of aluminum are distributed in a mixture of silicon and germanium (aluminum silicon germanium composite structure), a drain region 94, a gate insulating region 95, a gate electrode 96 and a source region 97.

As aluminum is used as quantum dots as shown in FIG. 9, the electric characteristics of the channel part can be controlled by using the influence of the electric charge accumulated in the quantum dots. Additionally, an electric charge can be accumulated in quantum dots for a long time, a nonvolatile memory from which the stored information is not lost after turning off the power supply can be formed by using such quantum dots. While an aluminum silicon germanium structure is used in the above description, other materials may be used so long as a structure comprising distributed pillar-shaped members can be obtained (e.g., aluminum silicon may be used in place of aluminum silicon germanium).

A single-electron memory that utilizes the quantum effect, or a single-electron transistor utilizing a similar principle can be formed by using an aluminum silicon germanium structure prepared in a manner as described above.

According to the invention, aluminum pillar-

shaped nano-structures such as quantum dots or quantum wires can be utilized in various different ways to remarkably broaden the scope of utilization of such structures.

5 (Example 1: first material Al, second material Si)

FIG. 3 is a schematic perspective view of an aluminum silicon structure according to the invention. In FIG. 3, the pillar-shaped members surrounded by silicon are aluminum structures, or  
10 aluminum wires having a diameter  $2r$  of 3 nm and a length  $L$  of 200 nm. Any two adjacently located pillar-shaped members are separated by a gap  $2R$  of 7 nm. In FIG. 3, reference symbols 21, 22 and 23 respectively denote a substrate, an aluminum  
15 quantum wire and silicon. An actual SEM image is shown in FIG. 6.

Firstly, the process used for preparing the aluminum wires will be described below.

An aluminum silicon composite film containing  
20 silicon by 55 atomic% relative to the total amount of silicon and aluminum was formed to a thickness of 200 nm on a glass substrate 21 by means of RF magnetron sputtering. A total of eight 15 mm square silicon chips 13 were placed on a 4-inch  
25 aluminum target as shown in FIG. 2 and used as so many targets. The sputtering operation was conducted by using an RF power source under

sputtering conditions of Ar gas flow rate: 50 sccm, discharge pressure: 0.7 Pa and applied power: 1 kW. The substrate was held to room temperature.

While eight silicon chips 13 were placed on  
5 an aluminum target and used as so many targets 12  
in this example, the number of silicon chips is by  
no means limited thereto and any other number of  
silicon chips may be used so long as the obtained  
aluminum silicon composite film contains silicon by  
10 about 55 atomic%. Additionally, it is not  
necessary to place targets of silicon chips on an  
aluminum target. Inversely, aluminum chips may be  
placed on a silicon target. Alternatively, targets  
prepared by baking powder of aluminum and silicon  
15 may be used.

Thereafter, the obtained aluminum silicon  
composite film was analyzed by means of ICP  
(inductively coupled plasma atomic emission  
spectrometry) in order to determine the amount of  
20 silicon relative to the total amount of aluminum  
and silicon (in terms of atomic%). As a result, it  
was found that the amount of silicon relative to  
the total amount of aluminum and silicon was about  
55 atomic%. For the convenience of analysis, a  
25 carbon substrate was used and the aluminum silicon  
composite film was formed on the carbon substrate.

Then, the aluminum silicon composite film was

observed through an FE-SEM (field emission scanning electron microscope). When observed from a position right above the substrate, the surface of the specimen showed circular plan views of aluminum nano-structures that were arranged two-dimensionally and surrounded by silicon. The circular plan views of the aluminum nano-structures had a diameter of 3 nm and the centers of any two adjacently located ones are separated by 7 nm on the average. When the specimen was observed through the FE-SEM along a cross section taken through the substrate, the aluminum nano-structures showed a height of 200 nm. It was found that the aluminum nano-structures were independent from each other.

When the specimen was observed by means of X-ray diffractometry, no silicon peak that reveals crystallinity was observed to prove that the silicon was amorphous. Some aluminum peaks, on the other hand, were observed to prove that the aluminum was at least partly polycrystalline.

Thus, it was possible to prepare an aluminum silicon nano-structure comprising aluminum wires having a diameter  $2r$  of 3 nm and a length  $L$  of 200 nm and surrounded by silicon in such a way that any two adjacently located aluminum wires were separated from each other by a gap  $2R$  of 7 nm.

As described above for the example, it is possible to form an aluminum silicon nano-structure comprising aluminum nano-structures, which may be aluminum quantum dots or aluminum quantum wires of dimensions of several nm, in the silicon matrix on the surface of a substrate by means of a film forming method such as a sputtering method that is adapted to form a film of substances in a non-equilibrium state.

10 (Example for Comparison)

An aluminum silicon composite film containing silicon by 15 atomic% relative to the total amount of silicon and aluminum was formed to a thickness of about 200 nm on a glass substrate by means of sputtering to prepare Specimen A for comparison. A pair of 15 mm square silicon chips<sup>13</sup> were placed on a 4-inch aluminum target and used as so many targets. The sputtering operation was conducted by using an RF power source under sputtering conditions of Ar gas flow rate: 50 sccm, discharge pressure: 0.7 Pa and applied power: 1 kW. The substrate was held to room temperature.

Thereafter, the obtained Specimen A for comparison was observed through an FE-SEM (field emission scanning electron microscope). See Fig. 7. When observed from a position right above the substrate, the surface of the specimen did not show

circular plan views of aluminum nano-structures. They showed plan views of aluminum connected continuously. In other words, the specimen was not a microstructure comprising pillar-shaped aluminum structures uniformly distributed in a silicon region. Their sizes by far exceeded 10 nm. When the specimen was observed through the FE-SEM along a cross section taken through the substrate, the aluminum portions showed a width exceeding 15 nm. The obtained aluminum silicon composite film was analyzed by means of ICP (inductively coupled plasma atomic emission spectrometry) in order to determine the amount of silicon relative to the total amount of aluminum and silicon (in terms of atomic%). As a result, it was found that the amount of silicon relative to the total amount of aluminum and silicon was about 15 atomic%.

Another aluminum silicon composite film containing silicon by 75 atomic% relative to the total amount of silicon and aluminum was formed to a thickness of about 200 nm on a glass substrate by means of sputtering to prepare Specimen B for comparison. A total of fourteen 15 mm square silicon chips 13 were placed on a 4-inch aluminum target and used as so many targets. The sputtering operation was conducted by using an RF power source under sputtering conditions of Ar gas flow rate: 50

sccm, discharge pressure: 0.7 Pa and applied power: 1 kW. The substrate was held to room temperature.

Thereafter, the obtained Specimen B for comparison was observed through an FE-SEM (field emission scanning electron microscope). When observed from a position right above the substrate, no aluminum parts were found. No aluminum parts were found either when the specimen was observed through the FE-SEM along a cross section taken through the substrate. The obtained aluminum silicon composite film was analyzed by means of ICP (inductively coupled plasma atomic emission spectrometry) in order to determine the amount of silicon relative to the total amount of aluminum and silicon (in terms of atomic%). As a result, it was found that the amount of silicon relative to the total amount of aluminum and silicon was about 75 atomic%.

Still eight other specimens were prepared for comparison under the conditions same as those used for preparing the Specimen A for comparison except that the number of silicon chips was changed. The prepared specimens contained silicon respectively by 20 atomic%, 25 atomic%, 35 atomic%, 50 atomic%, 55 atomic%, 60 atomic%, 65 atomic% and 70 atomic%. Table 1 below shows those having a microstructure where pillar-shaped aluminum structures were



distributed in a silicon region (formed) and those having no such a microstructure (not formed). Preferable range of silicon content is not less than 30 atomic% and not more than 60 atomic% from the viewpoint of uniformity of pillar-shaped structures. The specimens with a silicon content of 65 or 70 atomic% showed a lower crystallinity of aluminum, close to amorphous.

Table 1

silicon content (atomic%)	microstructure
15 (Specimen A for comparison)	not formed
20	formed
25	formed
35	formed
50	formed
55	formed
60	formed
65	Formed
70	Formed
75 (Specimen B for comparison)	not formed

10

Thus, structures comprising aluminum pillar-shaped structures distributed in a silicon region can be realized by regulating the content of

silicon to not less than 20 atomic% and not more than 70 atomic% relative to the total amount of aluminum and silicon. Further, the diameter (major axis) of the aluminum nano-structures of the microstructure to be prepared can be controlled by regulating the content ratio of aluminum to silicon. Then, it is also possible to obtain highly linear aluminum wires. A TEM (transmission electron microscope) may be used in place of an SEM to observe the structures.

Still another aluminum silicon composite film containing silicon by 55 atomic% relative to the total amount of silicon and aluminum was formed to a thickness of about 200 nm on a glass substrate by means of sputtering to prepare Specimen C for comparison. A total of eight 15 mm square silicon chips 13 were placed on a 4-inch aluminum target and used as so many targets. The sputtering operation was conducted by using an RF power source under sputtering conditions of Ar gas flow rate: 50 sccm, discharge pressure: 0.7 Pa and applied power: 1 kW. The substrate was held to 350°C.

Thereafter, the obtained Specimen C for comparison was observed through an FE-SEM (field emission scanning electron microscope). When observed from a position right above the substrate, a large lump of aluminum was found. Further, in a

X-ray diffraction measurement, crystallization of silicon was noted. In other words, no visible aluminum nano-structures were found and the silicon region was not amorphous but crystallized. The  
5 inventors presume that a transition to a more stable state occurs when the temperature of the substrate is too high so that no film growth takes forming aluminum nano-structure place..

It is also preferable to use a target  
10 composition of Al : Si = 55 : 45 for obtaining a structure where pillar-shaped members are uniformly distributed.

(Example 2)

FIG. 4 is a schematic perspective view of  
15 still another aluminum silicon nano-structure according to the invention. The illustrated aluminum nano-structure parts are pillar-shaped and surrounded by silicon. They are aluminum quantum dots having a diameter  $2r$  of 6nm and a length  $L$  of  
20 10 nm, any two adjacently located ones of the aluminum quantum dots being separated by a gap  $2R$  of 9 nm. In FIG. 4, reference symbols 31, 32 and 33 respectively denote a substrate, an aluminum quantum dot and silicon. Firstly, the method used  
25 in this example for preparing an aluminum silicon nano-structure having aluminum nano-structure parts that were aluminum quantum dots will be described.

An aluminum silicon composite film containing silicon by 30 atomic% relative to the total amount of silicon and aluminum was formed to a thickness of about 10 nm on a silicon substrate by means of an electron beam evaporation method that is a physical evaporation method. More specifically, a crucible of silicon and that of aluminum were used simultaneously for electron beam evaporation.

Thereafter, the obtained aluminum silicon composite film was analyzed by means of ICP (inductively coupled plasma atomic emission spectrometry) in order to determine the amount of silicon relative to the total amount of aluminum and silicon (in terms of atomic%). As a result, it was found that the amount of silicon relative to the total amount of aluminum and silicon was about 30 atomic%. For the convenience of analysis, a carbon substrate was used and the aluminum silicon composite film was formed on the carbon substrate.

Then, the aluminum silicon composite film was observed through an FE-SEM (field emission scanning electron microscope). When observed from a position right above the substrate, the surface of the specimen showed circular plan views of aluminum quantum dots that were arranged two-dimensionally and surrounded by silicon as shown in FIG. 4. The circular plan views of the aluminum nano-structures

(aluminum quantum dots) had a diameter  $2r$  of 6 nm and the centers of any two adjacently located ones are separated by a gap  $2R$  of 9 nm on the average. When the specimen was observed through the FE-SEM  
5 along a cross section taken through the substrate, the aluminum nano-structures showed a height of 10 nm. It was found that the aluminum quantum dots were independent from each other.

Thus, it was possible to prepare an aluminum  
10 silicon nano-structure comprising aluminum quantum dots having a diameter  $2r$  of 6 nm and a height  $L$  of 10 nm and surrounded by silicon in such a way that any two adjacently located aluminum dots were separated from each other by a gap  $2R$  of 9 nm.  
15 When the specimen was observed by means of X-ray diffractometry, it was found that the aluminum was crystalline and the silicon was amorphous.

(Example 3)

In this example, a specimen was prepared by  
20 using aluminum as first and silicon and germanium as second material. This example will be described by referring also to FIG. 3. FIG. 3 is a schematic perspective view of an aluminum silicon germanium composite structure comprising aluminum nano-  
25 structures having a pillar-shaped profile with a diameter  $2r$  of 7 nm and a length  $L$  of 200 nm and surrounded by silicon germanium, the gap  $2R$

separating any two adjacently located aluminum nano-structures being 10 nm. In FIG. 3, reference symbols 21, 22 and 23 respectively denote a substrate, an aluminum quantum wire (a pillar-shaped structure formed to contain aluminum) and an amorphous silicon germanium region. An SEM image is shown in FIG. 10.

Firstly, the process used for preparing the aluminum wires will be described below.

10       An aluminum silicon germanium composite film, whose content of silicon and germanium was 38 atomic% relative to the total amount of aluminum, silicon and germanium, was formed to a thickness of about 200 nm on a glass substrate 21 by means of RF magnetron sputtering. A pair of 15 mm square silicon chips and a pair of 15 mm square germanium chips were placed on a 4-inch aluminum target as shown in FIG. 2 and used as so many targets. The sputtering operation was conducted by using an RF power source under sputtering conditions of Ar gas flow rate: 50 sccm, discharge pressure: 0.7 Pa and applied power: 300 W. The substrate was held to room temperature.

25       While four chips 13 including silicon chips and germanium chips were placed on an aluminum target and used as so many targets in this example, the number of silicon and germanium chips is by no

means limited thereto and any other number of silicon and germanium chips may be used so long as the obtained aluminum silicon germanium composite film contains silicon and germanium by about 38  
5 atomic% relative to the total amount of aluminum, silicon and germanium. Additionally, it is not necessary to place targets of silicon and germanium chips on an aluminum target. Alternatively, aluminum chips and germanium chips may be placed on  
10 a silicon target. Still alternatively, targets prepared by sintering powder of silicon, germanium and aluminum may be used.

While an RF sputtering method was used for the sputtering process in this example, methods  
15 that can be used for the sputtering process is not limited thereto. An ECR sputtering method, a DC sputtering method or an ion beam sputtering method may alternatively be used. The sputtering conditions are dependent on the sputtering system  
20 that is used for the sputtering process and therefore they are not limited to those listed above.

Thereafter, the obtained aluminum silicon germanium composite film was analyzed by means of  
25 ICP (inductively coupled plasma atomic emission spectrometry) in order to determine the combined content of silicon and germanium relative to the

total amount of aluminum, silicon and germanium (in terms of atomic%). As a result, it was found that the combined content of silicon and germanium relative to the total amount of aluminum, silicon and germanium was about 38 atomic%.

For the convenience of analysis, a carbon substrate was used and the aluminum silicon germanium composite film was formed on the carbon substrate.

Then, the aluminum silicon germanium composite film was observed through an FE-SEM (field emission scanning electron microscope). When observed from a position right above the substrate, the surface of the specimen showed circular plan views of aluminum nano-structures that were arranged two-dimensionally and surrounded by a mixture of silicon and germanium. The circular plan views of the aluminum nano-structures had a diameter of 7 nm and the centers of any two adjacently located ones are separated by 10 nm on the average as determined by processing the image of the aluminum nano-structures. When the specimen was observed through the FE-SEM along a cross section taken through the substrate, the aluminum nano-structures showed a height of 200 nm. It was found that the aluminum nano-structures were independent from each other.



When the specimen was observed by means of X-ray diffractometry, no silicon and germanium peaks that reveal crystallinity were observed to prove that the mixture of silicon and germanium was  
5 amorphous. On the other hand, aluminum peaks were observed to prove that aluminum was polycrystalline.

Thus, it was possible to prepare an aluminum silicon germanium composite structure comprising aluminum wires having a diameter  $2r$  of 7 nm and a  
10 length  $L$  of 200 nm and surrounded by a mixture of silicon and germanium in such a way that any two adjacently located aluminum wires were separated from each other by a gap  $2R$  of 10 nm.

As described above for the example, it is  
15 possible to two-dimensionally form an aluminum silicon germanium composite structure comprising aluminum nano-structures, which may be aluminum quantum dots or aluminum quantum wires of dimensions of several nm to tens of several nm, on  
20 the surface of a substrate by means of a film forming method such as a sputtering method that is adapted to form a film of substances in a non-equilibrium state.

(Example for Comparison)

25 An aluminum silicon germanium composite film, whose content of silicon and germanium was 15 atomic% relative to the total amount of aluminum,

silicon and germanium, was formed to a thickness of about 200 nm on a glass substrate by means of sputtering to prepare Specimen D for comparison. A 15 mm square silicon chip and a 15 mm square germanium chip were placed on a 4-inch aluminum target and used as so many targets. The sputtering operation was conducted by using an RF power source under sputtering conditions of Ar gas flow rate: 50 sccm, discharge pressure: 0.7 Pa and applied power: 300 W. The substrate was held to room temperature.

Thereafter, the obtained Specimen D for comparison was observed through an FE-SEM (field emission scanning electron microscope). When observed from a position right above the substrate, the surface of the specimen did not show circular plan views of aluminum nano-structures. The aluminum parts showed plan views that were connected continuously. In other words, the specimen was not a microstructure comprising pillar-shaped aluminum structures uniformly distributed in a silicon germanium region. Their sizes (lengths) by far exceeded 20 nm. When the specimen was observed through the FE-SEM along a cross section taken through the substrate, the aluminum parts were not pillar-shaped but formed a lump with a size exceeding tens of several nm. The obtained aluminum silicon germanium composite film

was analyzed by means of ICP (inductively coupled plasma atomic emission spectrometry) in order to determine the combined content of silicon and germanium relative to the total amount of aluminum, silicon and germanium (in terms of atomic%). As a result, it was found that the amount of silicon and germanium relative to the total amount of aluminum, silicon and germanium was about 15 atomic%.

Another aluminum silicon germanium composite film, whose content of silicon and germanium was 75 atomic% relative to the total amount of aluminum, silicon and germanium, was formed to a thickness of about 200 nm on a glass substrate by means of sputtering to prepare Specimen E for comparison. Five 15 mm square silicon chips and five 15 mm square germanium chips were placed on a 4-inch aluminum target and used as so many targets. The sputtering operation was conducted by using an RF power source under sputtering conditions of Ar gas flow rate: 50 sccm, discharge pressure: 0.7 Pa and applied power: 300 W. The substrate was held to room temperature.

Thereafter, the obtained Specimen E for comparison was observed through an FE-SEM (field emission scanning electron microscope). When observed from a position right above the substrate, no aluminum parts were found. No distinct aluminum

parts were found either when the specimen was observed through the FE-SEM along a cross section taken through the substrate. The obtained aluminum silicon germanium composite film was analyzed by means of ICP (inductively coupled plasma atomic emission spectrometry) in order to determine the combined content of silicon and germanium relative to the total amount of aluminum, silicon and germanium (in terms of atomic%). As a result, it was found that the amount of silicon and germanium relative to the total amount of aluminum, silicon and germanium (or 100 atomic%) was about 75 atomic%.

Thus, it is possible to form aluminum nanostructures in silicon germanium by regulating the content of silicon and germanium to not less than 20 atomic% and not more than 70 atomic% relative to the total amount of aluminum, silicon and germanium. Then, it is also possible to obtain highly linear aluminum wires.

Another aluminum silicon germanium composite film, whose content of silicon and germanium was 38 atomic% relative to the total amount of aluminum, silicon and germanium, was formed to a thickness of about 200 nm on a glass substrate by means of sputtering to prepare Specimen F for comparison. A pair of 15 mm square silicon chips and a pair of 15 mm square germanium chips were placed on a 4-inch

aluminum target and used as so many targets. The sputtering operation was conducted by using an RF power source under sputtering conditions of Ar gas flow rate: 50 sccm, discharge pressure: 0.7 Pa and applied power: 300 W. The substrate was held to 350°C.

Thereafter, the obtained Specimen F for comparison was observed through an FE-SEM (field emission scanning electron microscope). When observed from a position right above the substrate, no circular or elliptic aluminum areas were found. In other words, no visible aluminum nano-structures were found. The inventors presume that a transition to a more stable state occurs when the temperature of the substrate is too high so that no film growth forming pillar-shaped aluminum nano-structures as shown in FIGS. 1A and 1B or in FIG. 3 takes place.

(Example 4)

In this example, a specimen of aluminum silicon germanium composite structure was prepared. This specimen will be described by referring also to FIG. 4. The illustrated aluminum nano-structure parts are pillar-shaped and surrounded by silicon germanium. They are aluminum quantum dots having a diameter  $2r$  of 6nm and a length  $L$  of 10 nm, any two adjacently located ones of the aluminum quantum

dots being separated by a gap  $2R$  of 8 nm. In FIG. 4, reference symbols 31, 32 and 33 respectively denote a substrate, an aluminum quantum dot and an amorphous silicon germanium region. Firstly, the method used in this example for preparing an aluminum silicon germanium composite structure having pillar-shaped aluminum nano-structure parts that were aluminum quantum dots will be described. An aluminum silicon germanium composite film containing silicon and germanium by 33 atomic% relative to the total amount of aluminum, silicon and germanium was formed to a thickness of about 10 nm on a silicon substrate by means of a magnetron sputtering method.

Thereafter, the obtained aluminum silicon germanium composite film was analyzed by means of ICP (inductively coupled plasma atomic emission spectrometry) in order to determine the combined content of silicon and germanium relative to the total amount of aluminum, silicon and germanium (in terms of atomic%). As a result, it was found that the content of silicon and germanium relative to the total amount of aluminum, silicon and germanium was about 33 atomic%. For the convenience of analysis, a carbon substrate was used and the aluminum silicon germanium composite film was formed on the carbon substrate.

Then, the aluminum silicon germanium composite film was observed through an FE-SEM (field emission scanning electron microscope). When observed from a position obliquely above the substrate, the surface of the specimen showed circular plan views of aluminum quantum dots that were arranged two-dimensionally and surrounded by silicon germanium as shown in FIG. 4. The circular plan views of the aluminum quantum dots had a diameter  $2r$  of 6 nm and the centers of any two adjacently located ones are separated by a gap  $2R$  of 8 nm. When the specimen was observed through the FE-SEM along a cross section taken through the substrate, the aluminum quantum dots showed a height of 10 nm. It was found that the aluminum quantum dots were independent from each other.

Thus, it was possible to prepare an aluminum silicon germanium composite structure comprising aluminum quantum dots having a diameter  $2r$  of 6 nm and a height  $L$  of 10 nm and surrounded by silicon germanium in such a way that any two adjacently located aluminum quantum dots were separated from each other by a gap  $2R$  of 8 nm. When the specimen was observed by means of X-ray diffractometry, it was found that the aluminum was crystalline and the silicon germanium was amorphous.

The diameter (major axis) of the aluminum

nano-structures and the gap separating any two adjacently located aluminum nano-structures can be regulated by changing the content of silicon and germanium relative to the total amount of aluminum, silicon and germanium of an aluminum silicon germanium composite structure (typically, the diameter (major axis) of the aluminum nano-structures is increased by reducing the content of silicon and germanium). The height of the aluminum nano-structures of an aluminum silicon germanium composite structure can be regulated by changing the film thickness of the latter.

Further, the diameter of or gaps separating the pillar-shaped aluminum structures can be regulated by changing the content ratio of germanium to silicon in the aluminum silicon germanium composite structure.

As described in detail, the present invention provides a structure comprising pillar-shaped members distributed in a surrounding region, a method of manufacturing such a structure and a device realized by using such a structure.



## CLAIMS

1. A structure containing a first material and a second material, said structure comprising pillar-shaped members containing the first material and a region containing the second material and surrounding the pillar-shaped members provided that the second material is not composed of only germanium, the structure containing the second material at a content not less than 20 atomic% and not more than 70 atomic% relative to the total amount of the first and second materials.

2. The structure according to claim 1, wherein said first material is aluminum.

15

3. The structure according to claim 1 or 2, wherein said second material is silicon or a mixture of silicon and germanium.

4. The structure according to claim 1 or 2, wherein said structure contains said second material at a content not less than 30 atomic% and not more than 60 atomic%.

5. The structure according to claim 1, wherein the average diameter of said pillar-shaped members is not more than 50 nm.

6. The structure according to claim 1,  
wherein the distance between the centers of any two  
adjacent located ones of said pillar-shaped  
members contained in said structure is not more  
5 than 30 nm.

7. A device comprising a structure according  
to claim 1.

10 8. A method of manufacturing a structure,  
comprising a step of preparing a substrate and a  
step of forming a film on the substrate in a non-  
equilibrium state, the film having a structure  
comprising pillar-shaped members containing a first  
15 material and a region containing a second material  
and surrounding the pillar-shaped members provided  
that the second material is not composed of only  
germanium, the structure containing the second  
material at a content not less than 20 atomic% and  
20 not more than 70 atomic% relative to the total  
amount of the first and second materials.

9. A film-shaped aluminum silicon composite  
structure, comprising pillar-shaped structures  
25 containing aluminum and a silicon region  
surrounding the pillar-shaped structures, the  
composite structure containing silicon at a

content not less than 20 atomic% and not more than 70 atomic%.

10. The composite structure according to  
5 claim 9, wherein the content of silicon in said composite structure is not less than 25 atomic% and not more than 65 atomic%.

11. The composite structure according to  
10 claim 10, wherein the content of silicon in said composite structure is not less than 30 atomic% and not more than 60 atomic%.

12. The composite structure according to any  
15 of claims 9 through 11, wherein the diameters of said pillar-shaped structures are not less than 0.5 nm and not more than 50 nm.

13. The composite structure according to  
20 claim 12, wherein the diameters of said pillar-shaped structures are not less than 0.5 nm and not more than 10 nm.

14. The composite structure according to any  
25 of claims 9 through 11, wherein the distance between the centers of any two adjacently located ones of said pillar-shaped structures is not more

than 30 nm.

15. The composite structure according to claim 14, wherein the distance between the centers  
5 of any two adjacently located ones of said pillar-shaped structures is not more than 15 nm.

16. The composite structure according to any of claims 9 through 11, wherein said silicon region  
10 contains amorphous silicon as principal ingredient.

17. The composite structure according to claim 16, wherein said composite structure is formed on an insulator.

15

18. An electronic device comprising an aluminum silicon composite structure according to claim 9.

20 19. A method of manufacturing an aluminum silicon composite structure, comprising preparing said composite structure by means of a method of forming a film on a substrate in a non-equilibrium state.

25

20. A method of manufacturing a film-shaped aluminum silicon composite structure, comprising a

step of preparing a substrate and a step of forming  
a film on the substrate in a non-equilibrium state,  
the formed film having an aluminum silicon  
composite structure comprising pillar-shaped  
5 structures containing aluminum and a silicon region  
surrounding the pillar-shaped structures, the  
composite structure containing silicon at a content  
not less than 20 atomic% and not more than 70  
atomic%.

10

21. The method according to claim 20, wherein  
said step of forming a film in a non-equilibrium  
state is carried out by conducting a sputtering  
method.

15

22. The method according to claim 21, wherein  
said sputtering method is conducted by using a  
target material separately including aluminum and  
silicon.

20

23. The method according to claim 21, wherein  
said sputtering method is conducted by keeping a  
substrate at temperatures not higher than 300°C  
when forming an aluminum silicon composite film  
25 containing silicon by 20 to 70 atomic% relative to  
the total amount of aluminum and silicon.

24. A film-shaped aluminum silicon germanium composite structure, comprising pillar-shaped structures containing aluminum and a silicon germanium region surrounding the pillar-shaped structures, the composite structure containing silicon and germanium at a combined content not less than 20 atomic% and not more than 70 atomic%.

25. The composite structure according to claim 24, wherein the combined content of silicon and germanium in said composite structure is not less than 25 atomic% and not more than 65 atomic%.

26. The composite structure according to claim 25, wherein the combined content of silicon and germanium in said composite structure is not less than 30 atomic% and not more than 60 atomic%.

27. The composite structure according to any of claims 24 through 26, wherein the diameters of said pillar-shaped structures are not less than 0.5 nm and not more than 30 nm.

28. The composite structure according to claim 27, wherein the diameters of said pillar-shaped structures are not less than 0.5 nm and not more than 15 nm.

29. The composite structure according to any  
of claims 24 through 26, wherein the distance  
between the centers of any two adjacently located  
ones of said pillar-shaped structures is not more  
5 than 30 nm.

30. The composite structure according to  
claim 29, wherein the distance between the centers  
of any two adjacently located ones of said pillar-  
10 shaped structures is not more than 20 nm.

31. The composite structure according to any  
of claims 24 through 26, wherein said silicon  
germanium region contains amorphous silicon  
15 germanium as principal ingredient.

32. The composite structure according to any  
of claims 24 through 26, wherein if the content  
ratio of silicon to germanium in said silicon  
20 germanium region is expressed as composition  $\text{Si}_x\text{Ge}_{1-x}$   
where Si represents silicon and Ge represents  
germanium,  $x$  is within the range of  $0 < x < 1$ .

33. The composite structure according to  
25 claim 31, wherein said composite structure is  
formed on an insulator.

34. An electronic device comprising an aluminum silicon germanium composite structure according to claim 24.

5           35. A method of manufacturing an aluminum silicon germanium composite structure, comprising preparing said composite structure by means of a method of forming a film on a substrate in a non-equilibrium state.

10

          36. A method of manufacturing a film-shaped aluminum silicon germanium composite structure, comprising a step of preparing a substrate and a step of forming a film on the substrate in a non-equilibrium state, the formed film having an  
15           aluminum silicon germanium composite structure comprising pillar-shaped structures containing aluminum and a silicon germanium region surrounding the pillar-shaped structures, the composite  
20           structure containing silicon and germanium at a combined content not less than 20 atomic% and not more than 70 atomic%.

          37. The method according to claim 36, wherein  
25           said step of forming a film in a non-equilibrium state is carried out by conducting a sputtering method.

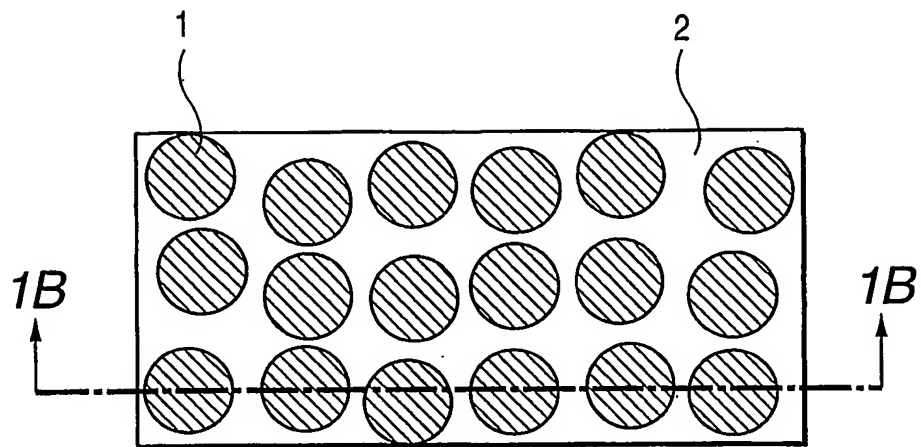
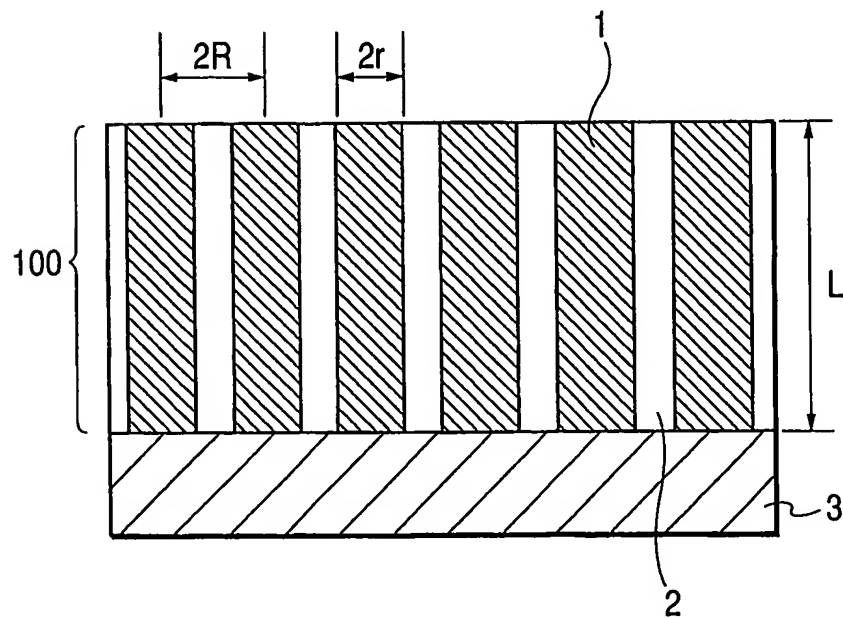


38. The method according to claim 37, wherein said sputtering method is conducted by keeping a substrate at temperatures not higher than 300°C when forming an aluminum silicon germanium composite film containing silicon and germanium by 20 to 70 atomic% relative to the total amount of aluminum, silicon and germanium.

39. A structure comprising a plurality of pillar-shaped members and a region surrounding the lateral surfaces of the plurality of pillar-shaped members, the pillar-shaped members having a diameter not more than 20 nm, the distance between the centers of any two adjacently located ones of the pillar-shaped members being not more than 30 nm.

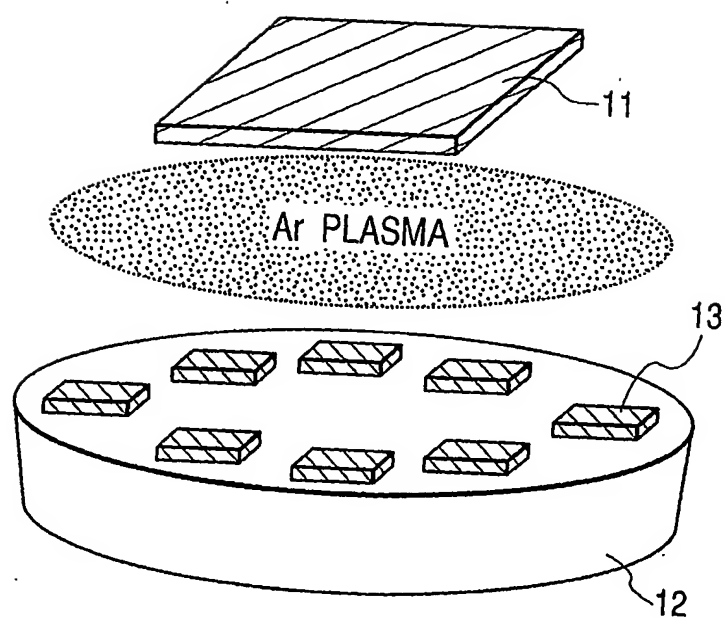
40. A structure comprising pillar-shaped members containing aluminum and a region surrounding the pillar-shaped members, the pillar-shaped members and the region being formed simultaneously, the pillar-shaped members having a diameter not more than 20 nm.

1/8

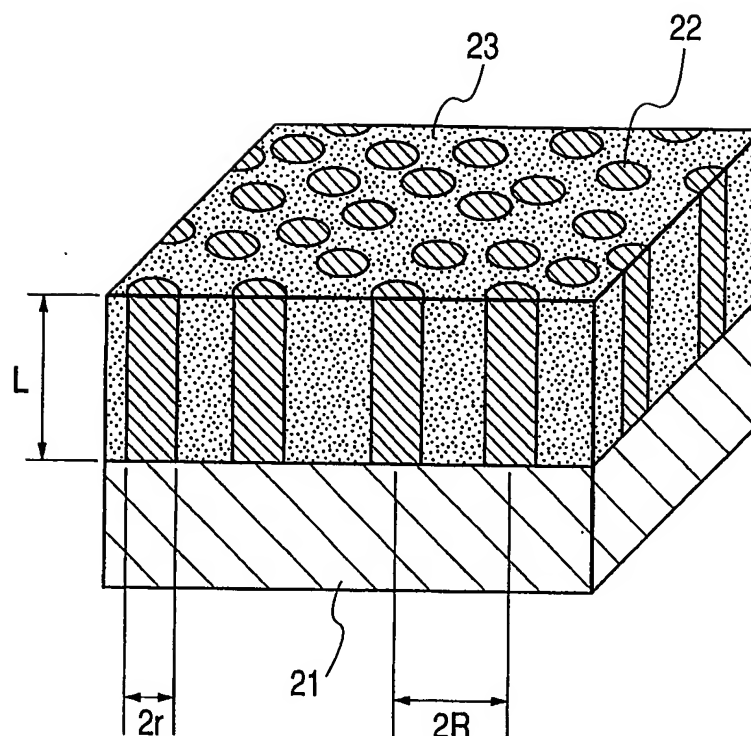
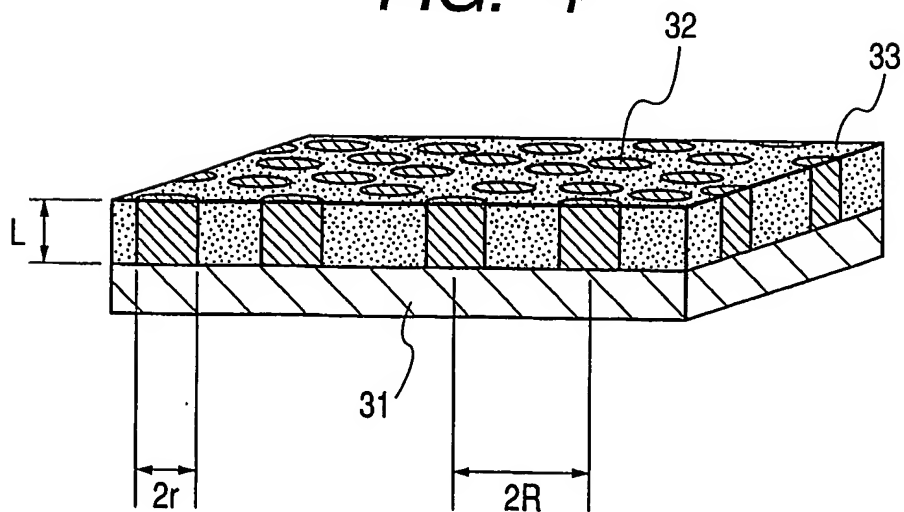
*FIG. 1A**FIG. 1B*

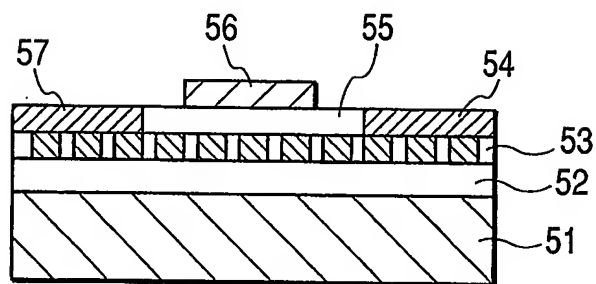
2/8

FIG. 2

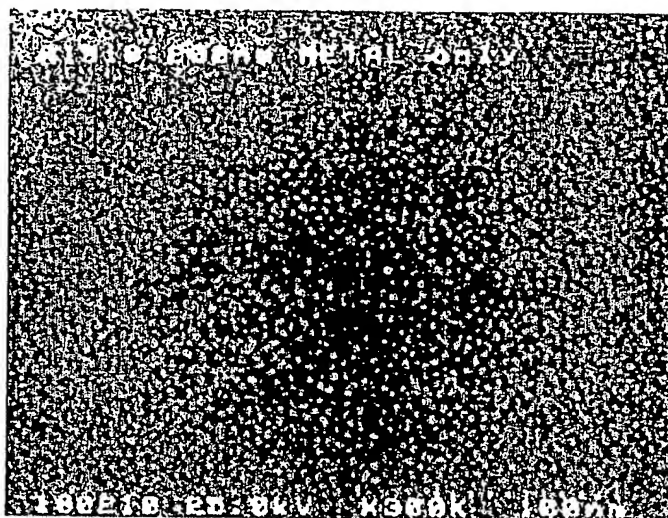
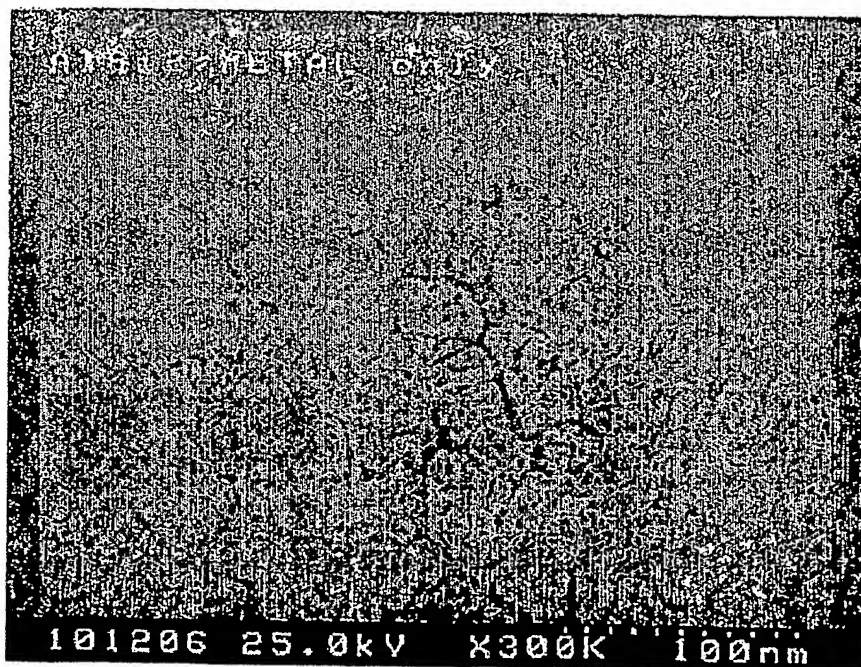


3/8

**FIG. 3****FIG. 4**

*FIG. 5*

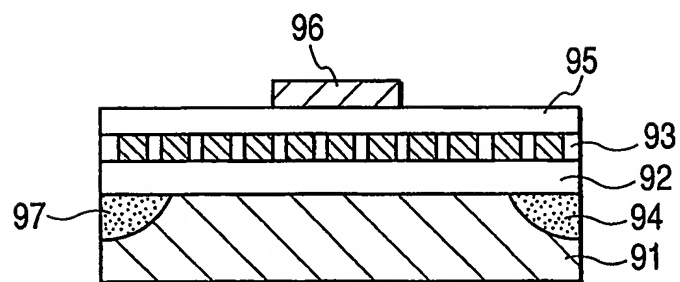
5/8

*FIG. 6**FIG. 7*

6/8

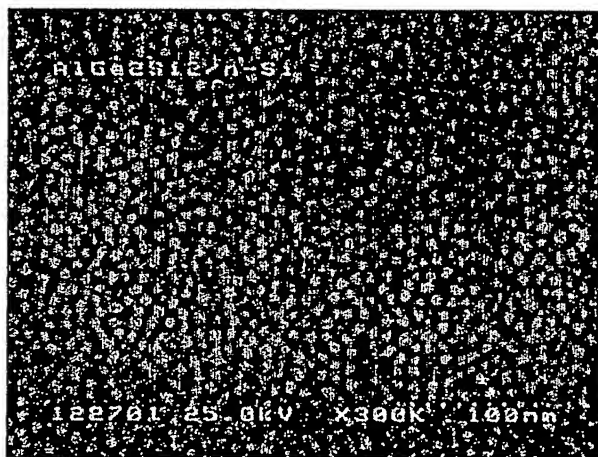
*FIG. 8*



**FIG. 9**



*FIG. 10*



## INTERNATIONAL SEARCH REPORT

International Application No

PCT/JP 03/01368

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 H01L29/12 H01L21/203 B82B1/00 B82B3/00

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 H01L B82B H01S

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the International search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ, WPI Data, INSPEC

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2001/042502 A1 (LEE SI-CHEN ET AL) 22. November 2001 (2001-11-22) the whole document ---	1-40
A	EP 0 851 506 A (SANYO ELECTRIC CO) 1 July 1998 (1998-07-01) the whole document ---	1-40
A	US 5 369 873 A (WALTERS COLIN R ET AL) 6 December 1994 (1994-12-06) the whole document ---	1-40
A	US 5 240 558 A (KAWASAKI HISAO ET AL) 31. August 1993 (1993-08-31) the whole document ---	1-40
	--- -/--	

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents:

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

22 May 2003

Date of mailing of the international search report

11.06.2003

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

PÄR MORITZ /EÖ

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/JP 03/01368

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	PATENT ABSTRACTS OF JAPAN vol. 1999, no. 14, 22 December 1999 (1999-12-22) & JP 11 251334 A (FURUKAWA ELECTRIC CO LTD:THE), 17 September 1999 (1999-09-17) abstract ---	1-40
A	PATENT ABSTRACTS OF JAPAN vol. 1999, no. 05, 31 May 1999 (1999-05-31) & JP 11 031862 A (NEC CORP), 2 February 1999 (1999-02-02) abstract ---	1-40
A	PATENT ABSTRACTS OF JAPAN vol. 007, no. 255 (E-210), 12 November 1983 (1983-11-12) & JP 58 142523 A (TADASHI MIZOGUCHI), 24 August 1983 (1983-08-24) abstract -----	1-40

## INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/JP 03/01368

Patent document cited in search report		Publication date		Patent family member(s)	Publication date
US 2001042502	A1	22-11-2001	TW	447013 B	21-07-2001
EP 0851506	A	01-07-1998	JP	10189779 A	21-07-1998
			EP	0851506 A2	01-07-1998
US 5369873	A	06-12-1994	EP	0538338 A1	28-04-1993
			FI	930034 A	05-01-1993
			WO	9201316 A1	23-01-1992
			GB	2246727 A ,B	12-02-1992
			JP	6501810 T	24-02-1994
US 5240558	A	31-08-1993	NONE		
JP 11251334	A	17-09-1999	NONE		
JP 11031862	A	02-02-1999	JP	2993470 B2	20-12-1999
JP 58142523	A	24-08-1983	NONE		

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

**BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☐ FADED TEXT OR DRAWING
- ☒ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☐ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**